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Nanocomposite with Non-Spherical Granules – Logarithmic Field Dependence of Giant Magnetoresistance

E. Meilikhov[a], B. Raquet[b]
[a]Russian Research Center «Kurchatov Institute», 123182 Moscow, Russia
[b]Laboratoire de Physique de la Matière Condensée de Toulouse, Av. de Rangueil, 31077 Toulouse, France

ABSTRACT

Magnetoresistance of the nanocomposite $Fe_x(SiO_2)_{1-x}$ ($x \approx 0.6$) at high enough magnetic field is logarithmic function of the magnetic field. Such a dependence does not fall into the known theory of giant magnetoresistance of ferromagnetic nanocomposites. This paper examines the giant magnetoresistance of such a system in terms of a simple model where the non-ordinary quasi-logarithmic magnetic field dependence of nanocomposite magnetoresistance is related to the non-spherical granules' distribution over their shapes.

INTRODUCTION

In the present paper, we consider the giant magnetoresistance of the nanocomposite $Fe_x(SiO_2)_{1-x}$ which is the granular ferromagnetic metal in the insulator matrix with $x \approx 0.6$ (that corresponds to the metal state close to the percolation metal-insulator transition). Experiments show that at high enough magnetic fields, the resistance of the system depends logarithmically on the magnetic field. Such a dependence does not fall into the framework of the known theory of the giant magnetoresistance of ferromagnetic nanocomposites [1, 2]. We relate this discrepancy to the fact that the "traditional" theory is applied to systems with spherical granules, while real nanocomposites consist most commonly of non-spherical ones. Moreover, as a rule, there are granules of diverse non-sphericity in the system - from the strongly prolate to the strongly oblate ones. This paper examines the giant magnetoresistance of such a system in terms of a simple model where the non-ordinary semilogarithmic magnetic field dependence of nanocomposite magnetoresistance is related to the non-spherical granules' distribution over their shapes.

EXPERIMENT

Thin $Fe_x(SiO_2)_{1-x}$ films with $x \approx 0.6$ were studied. They have been prepared by the ion-beam sputtering technique in a vacuum chamber with a mosaic target consisting of Fe and SiO_2 tablets. The volume fraction x of iron has been controlled by X-ray micro-analysis. Grains' diameters vary from ~2 nm up to ~ 20 nm and the film thickness equals about 0.4 μm.

The magnetoresistance $\Delta R/R$ of the film (R is the sample resistance at a given temperature and zero magnetic field, ΔR is the resistance change at the magnetic field B) was measured within the 4.2-300 K temperature range under the action of the "long" (of ~0.1 s-duration) pulse magnetic fields up to 20 T.

Experimental field dependencies of the sample manetoresistance for various temperatures are shown in Fig. 1. In fig. 2, only those parts of those dependencies are demonstrated that relate to the high field region. In this case, the magnetoresistance is shown

as a function of the "effective" field B/T [1]. It could be seen that in the high field region the magnetoresistance is the logarithmic function of the magnetic field.

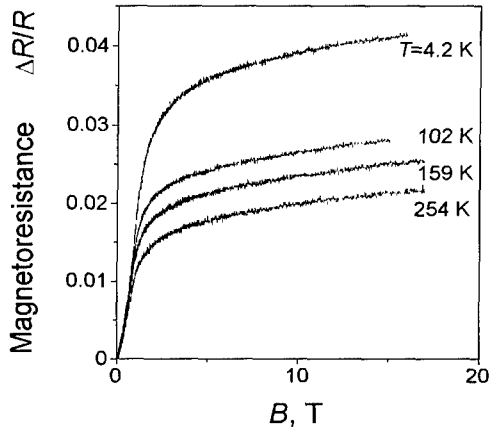

Fig. 1. Field dependencies of $Fe_x(SiO_2)_{1-x}$ nanocomposite magnetoresistance (x≈0.6) at various temperatures.

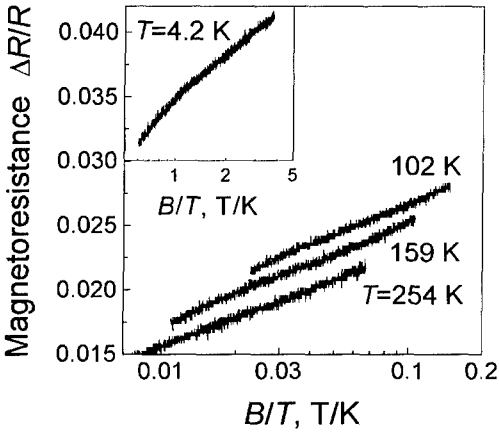

Fig.2. Log-scale field dependencies of $Fe_x(SiO_2)_{1-x}$ nanocomposite magnetoresistance (x≈0.6) at various temperatures in the high field region.

THEORY: NANOCOMPOSITE WITH NON-SPHERICAL GRANULES

The conductivity of granular metals (nanocomposites) and (in the case of ferromagnetic metals) their giant magnetoresistance are defined by tunnel intergranular electron transitions [1, 2]. However, in real systems consisting of granules of various sizes, not every

[1] It is known that for non-interacting spherical granules the magnetoresistance $\Delta R/R \propto M^2$, where M is the nanocomposite magnetization [2]. In the case, the latter is defined by the Langevin function (cf. Eq.(4)) which argument is the effective field B/T.

those transitions are actual: the most considerable contribution to the conductivity is due to tunnel transitions between granules of sizes close to the "optimal" one [3, 4]. For a nanocomposite consisting of *spherical* granules, such an optimal size is defined by the competition between enhanced concentration of small granules (that is typical for real systems) and reduced degree of their ionization which is the consequence of Coulomb effects. So, it is the result of the "game of survival" – there is great deal of small granules but only a few of them are charged (and therefore could contribute to the conductivity) and there is a few of big granules which all are charged. The optimal granule size is expressed by the relation [3]

$$a_{opt}(T) \approx a_0 (x/4\pi)^{1/2} (\lambda/a_0)(T/T_0)^{-5/9} \qquad (1)$$

where $kT_0 \approx (e^2/\varepsilon\, a_0)(a_0/\lambda)^{3/2} x^{-1/2}[1-(x/x_c)^{1/3}]$, a_0 is the average granule size, λ is the electron wavelength in insulator phase, ε is its dielectric constant, x is the volume fraction of the metal phase, and x_c is the percolation threshold. So, the conductance $G(T)$ of the system is controlled by the "optimal cluster" consisting of particles of the optimal size $a_{opt}(T)$ which is rearranged with temperature.

The resistance variation of the considered system under magnetic field B is defined by the magnetic field dependence of the probability of tunnel transitions between spontaneously magnetized single-domain (due to small sizes) granules. The reduced magnetoresistance $MR(B, T) = [G(0,T)-G(B,T)]/G(0,T)$ equals [4]

$$\Delta R(B,T)/R = P^2 \langle \cos\gamma \rangle^2 \qquad (2)$$

where P is the electron spin polarization in the ferromagnetic granule, γ are angles between the external magnetic field and magnetic moments of granules. Averaging is performed over the granules constituting the optimal cluster. Thus, the calculation of the magnetoresistance is reduced to the calculation of the averaged (over that cluster) $<\cos\gamma>$ - value.

However, in the real system, granules are, generally, *non-spherical*. It means that not every values of γ- angle for a single granule are equally probable and the averaged (over time) $\overline{\cos\gamma}$ - value for a non-spherical granule is defined by its magnetic anisotropy (crystal or geometric) and the external field [5]. For an ellipsoidal granule with a high (comparing to Bohr magneton) magnetic moment,

$$\overline{\cos\gamma} = \int_0^{\pi} \exp[-(W_A + W_B)/kT] \cos\gamma\, d\Omega \Big/ \int_0^{\pi} \exp[-(W_A + W_B)/kT]\, d\Omega, \qquad (3)$$

where $d\Omega = \sin\gamma\, d\gamma d\phi$ is the solid angle, ϕ is the azimuth angle of granule' magnetic moment ($\cos\phi = [\cos\theta - \cos\gamma\cos\beta]/\sin\gamma\sin\beta$, θ and β are the angles between the large axis of a granule and directions of its magnetic moment and magnetic field, respectively). In Eq. (3) $W_A = W_A(\theta)$ is the magnetic anisotropy energy independent of the magnetic field, $W_B = -I_s V B \cos\gamma$ is the Zeeman energy depending on the γ- angle only, I_s is the saturation magnetization of granule' material. At high magnetic fields, $W_B >> W_A$ and, hence,

$$\overline{\cos\gamma} = \mathrm{cth}\, h - 1/h \equiv L(h), \qquad (4)$$

where $h = I_s V B / kT$. That corresponds to the known Langevin model.

If the optimal cluster consisted of *spherical* granules of the size a_{opt}, then the granule volume V appearing in Eq. (4) would be the same for all granules and equal to $V = V_{opt} = (4\pi/3)\, a^3_{opt}(T) \propto T^{-5/3}$. In this case, $\langle\cos\gamma\rangle = \overline{\cos\gamma} = L(h_{opt})$, where $h_{opt} = I_s B V_{opt}/kT \propto T^{-8/3}$. It is clear that even in this case the temperature dependence of the optimal cluster magnetic moment (which is proportional to $<\cos\gamma>$) is not described by the Langevin model (where $h \propto 1/T$).

In the system consisting of *non-spherical* (ellipsoidal) granules, the situation is more complicate. In that case, the probability of granules' ionization is defined by their *capacity C* (which in the spherical case coincides with the granule radius). Therefore, the optimal granule size is the result of the new "game of survival" which depends on 1) the relation between the charged granules' concentration and their capacity, and 2) the dependence of the average (tunnel) distance between granules with equal capacities and their characteristic size. A granule which form is an ellipsoid of revolution has the two characteristic sizes a, b that are the lengths of its long and short axes. Which of those sizes is the characteristic one for the problem considered? It is known that the capacity of the ellipsoidal granule with the larger size of a depends slightly on its smaller size b: for the prolate ellipsoid of revolution $C=(a^2-b^2)^{1/2}$ /Arch$(a/b) \approx a/\ln(2a/b)$, while for the oblate one $C=(a^2-b^2)^{1/2}$ /arccos(a/b) (i.e., $2/\pi<C/a<1$) [6]. It means that the only essential size of an ellipsoidal granule is its larger size, and *all* granules with the larger size a close to a_{opt} are the optimal ones. Therefore, the optimal cluster consists of granules of various volumes in the range $V_{min} < V < V_{opt}$, where $V_{opt}= (4\pi/3)a^3_{opt}$ and $V_{min} = (b_{min}/a_{opt})^2 V_{opt}$ (for prolate ellipsoids) or $V_{min} = (b_{min}/a_{opt})V_{opt}$ (for oblate ellipsoids), b_{min}/a_{opt} is the minimum ratio of the respective granule' sizes which characterizes their maximum (for a given nanocomposite) elongation or flatness. If $b_{min}/a_{opt} \sim 0.1$, then the optimal cluster includes granules which volumes differ by ~100 times! Naturally, in this case, $\langle \cos\gamma \rangle \neq \overline{\cos\gamma}$ and one should perform averaging over all granules of the optimal cluster. Let $f_b(b)$ be the distribution function of the smaller sizes of the granules, and x_o is the volume fraction of the prolate granules. Then

$$\langle \cos\gamma \rangle = \int_{b=b_{min}}^{b=a_{opt}} \left\{(1-x_o)L\left[(ab^2/a^3_{opt})h_{opt}\right] + x_o L\left[(a^2 b/a^3_{opt})h_{opt}\right]\right\} f_b(b)db = F(h_{opt}),$$

$$F(h_{opt}) = \int_{z_{min}}^{1} \left[(1-x_o)L(h_{opt}z^2) + x_o L(h_{opt}z)\right] f_z(z)dz, \qquad (5)$$

where we introduced the distribution function $f_z(z)$ of the parameter $z \equiv b/a_{opt}$ ($0<z<1$, $z_{min}=b_{min}/a_{opt}$).

The distribution function $f_z(z)$ is, most likely, dependent on the technique of nanocomposite producing. This refers equally to the relation between numbers of prolate and oblate granules that is defined by the parameter x_o. In principle, the relevant information may be obtained by means of electron-microscopic investigations of the system in question. But, as calculations show (see below), the qualitative form of the magnetic field dependence of the magnetoresistance is not critical neither to choosing the distribution function $f_z(z)$, nor to values of the parameters $z_{min}<<1$ and x_o. So, as a simple approximation one could consider the system with the uniform distribution function ($f_z(z)$=const) consisting of prolate granules only (x_o=0), that is the system where prolate ellipsoidal granules of any form, from the spherical (z=1) to needle-like (z=0), are equally probable. In calculations, z_{min} =0.1 was accepted.

Magnetic field dependence of the magnetoresistance of such a system calculated with Eq. (5) is presented in Fig. 3 (the solid line 2). It could be seen that within the broad range of magnetic fields (in this case, at $5<h_{opt}<50$), the field dependence of the magnetoresistance is nearly logarithmic one. As calculations show (see Fig. 3) the character of that dependence is qualitatively the same for various functions $f_z(z)$ and x_o-values. What changes, it is the range of magnetic fields where that dependence is quasi-logarithmic one.

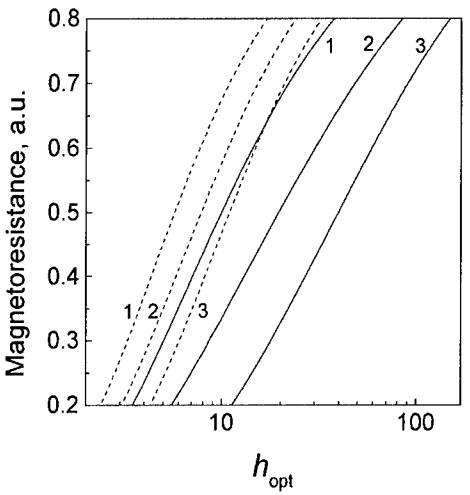

Fig.3. Calculated magnetic field dependencies of the magnetoresistance for the granular ferromagnetic metal with non-spherical granules of prolate (solid lines, x_0=0) and oblate (dashed lines, x_0=1) form. The distribution function $f_z(z)$: 1 - linearly-increasing, 2 - uniform, 3 - linearly-decreasing.

Characteristic effective field for the magnetic field range, where quasi-logarithmic dependence of the magnetoresistance should be observed, is of about h_{opt}~20 (cf. Fig. 3, the solid line 2 for the uniform distribution function $f_z(z)$). Comparing this value with the value of B/T~$3\cdot10^{-2}$ T/K, corresponding to the experimentally determined ranges of the magnetoresistance logarithmic dependence at temperatures T~100-250 K (cf. Fig. 2), the diameter of the optimal granules $2a_{opt}$~20 nm could be derived (in doing so, the saturation magnetization of Fe-granules was considered as being equal to that for the bulk iron, I_s≈0.2 T). It is in a reasonable agreement with the data of electron-microscopic analysis of the investigated nanocomposite.[2]

Let us discuss now the magnetoresistance temperature dependence in the range of its logarithmic magnetic field dependence. In the framework of the considered model, there is the only reason for such a dependence - the variation of the larger granule size a_{opt} of the optimal cluster with temperature. That dependence occurs to be the same as Eq.(1), $a_{opt}(T) \propto T^{-5/9}$. It means that the magnetoresistance depends only on that parameters' combination which defines the value of $h_{opt} \propto BV_{opt}/T \propto B/T^{8/3}$. Therefore, in the framework of our model there is a parametric magnetoresistance dependence which has the following form $MR=MR(B/T^{8/3})$. In other words, every value of the reduced magnetization $\Delta R/R$ as a function of the parameter $B/T^{8/3}$ has to fall on a single master curve. Experimental data presented in the respective form (cf. Fig. 4) support that theoretical prediction. Thus, the considered model provides qualitatively correct description of the experimental results concerning the magnetoresistance of the granular ferromagnetic metal $Fe_x(SiO_2)_{1-x}$ under high magnetic fields.

In conclusion, we have shown that the model of the nanocomposite with granules of various non-spherical forms leads to the quasi-logarithmic magnetic field dependence the magnetoresistance of such a system under high enough fields. This is, obviously, associated

[2] According to Eq. (1), $a_{opt}(T) \propto T^{-1/2}$ and, hence, the calculated optimal granule size at T=4.2 K equals $2a_{opt}$(4.2 K) ~ 100 nm. However, there are no such big granules in the real system and, hence, it is incorrect to employ the considered model at so low temperatures.

with the great volume scattering of granules constituting the optimal cluster. The magnetoresistance saturates together with the magnetization of this cluster, but with

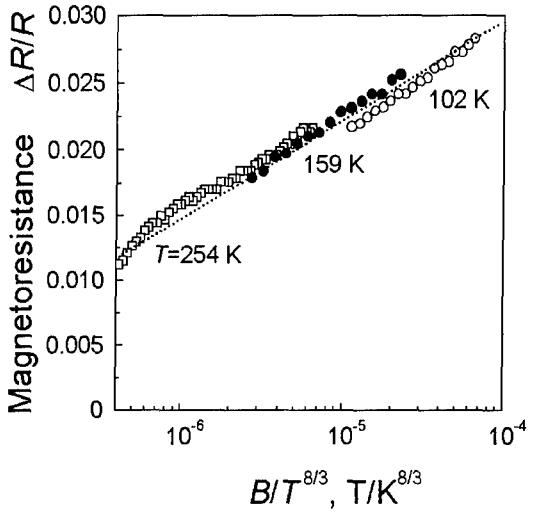

Fig. 4. Parametric dependencies of $Fe_x(SiO_2)_{1-x}$ – nanocomposite magnetoresistance (x≈0.6) in the high field region. The dotted master line is the guide for eye.

increasing field more and more small granules become contribute to the magnetization. It is this "extended" contribution that results in quite slow (quasi-logarithmic, as has been shown) saturation of the magnetization and, hence, of the magnetoresistance, as well.

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